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Research Progress on Mo-based Two-dimensional Materials in Non-enzymatic Glucose Sensors

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Abstract

This article simply reviews the research progress of non-enzymatic glucose sensors based on two-dimensional Mo-based materials (MoS_2, MoO_3, $MoSe_2$), with a focus on analyzing their electrocatalytic mechanisms, performance optimization strategies, and application potential. By comparing the sensitivity, detection limit, and stability of different materials, the advantages of molybdenum-based two-dimensional materials in sensor design are summarized. Furthermore, emerging applications in medical health, smart wearables, and environmental monitoring are discussed, and future research directions are proposed, including the development of multifunctional composite materials and breakthroughs in miniaturization technology. Finally, the future development of molybdenum-based non-enzymatic glucose sensors is prospected.

Keywords

Non-enzymatic Sensors; Two-dimensional Materials; Glucose Detection; Electrocatalysis; Mo-based Materials.

1. Introduction

Traditional blood glucose monitoring methods face issues of invasiveness and complicated procedures, making continuous real-time monitoring difficult. Traditional enzyme-based glucose sensors also suffer from high costs, poor stability, and susceptibility to environmental factors1[1]. To overcome these limitations, non-enzymatic glucose sensors have emerged, relying on the catalytic properties of materials. These sensors offer significant advantages in stability, cost, and environmental adaptability, enabling convenient real-time blood glucose monitoring[2]. They are of great significance for diabetes treatment, athlete training[3], and health management.

Molybdenum-based (Mo-based) non-enzymatic glucose sensors have gradually gained attention as an emerging research direction due to their low cost, high stability, good electrochemical performance, and broad application prospects[4]. Compared to non-enzymatic glucose sensors based on noble metals such as Au, Pt, and Ag, Mo-based materials like MoS_2, MoO_3, $MoSe_2$, and Mo-doped composites are relatively inexpensive[5], and can be produced on a large scale using simple chemical synthesis methods, thus reducing the overall cost of the sensor and enhancing its economic feasibility for large-scale applications[6]. Mo-based materials generally exhibit stronger corrosion resistance compared to metal nanomaterials. Particularly in strongly oxidative or acidic environments, metal nanomaterials may lose their catalytic activity due to surface oxidation or electrochemical corrosion, leading to a decline in sensor performance[7]. Compared to carbon-based materials such as carbon nanotubes and graphene, mo-based materials, especially MoS_2 and MoO_3, show better electrocatalytic activity in electrochemical catalytic processes. MoS_2, MoO_3, and $MoSe_2$ are all layered structure materials, where the atomic layers between each layer are spaced apart, allowing molecules or ions

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to diffuse and react through the interlayer gaps[8]. The layered structure provides these materials with greater flexibility in surface modification, doping, and compositing. By introducing metal atoms, oxides, or other elements between the layers, the electrochemical properties of the materials can be effectively adjusted, further enhancing their catalytic activity and selectivity[9]. Currently, there are relatively few reviews on the application of Mo-based materials in non-enzymatic glucose sensors. Zhu et al.[10]reviewed the latest advancements in non-enzymatic glucose sensors based on different metal oxides (such as ZnO, NiO, Co_3O_4, MnO_2, etc.) and their nanocomposites. Singh et al. [11]reviewed electrochemical sensors for glucose detection in human body fluids, focusing on the application of non-enzymatic glucose sensors for non-invasive detection.

Traditional blood glucose monitoring methods and enzyme-based glucose sensors have many drawbacks, which has led to the emergence of non-enzymatic glucose sensors. Mo-based non-enzymatic glucose sensors, as an emerging research direction, have attracted attention due to their advantages of low cost, high stability, and more. Compared to noble metal-based and carbon-based materials, molybdenum-based materials are cost-effective, corrosion-resistant, and exhibit good electrocatalytic activity, with their layered structure providing greater flexibility in surface modification. Currently, there are few reviews on their application research. This paper innovatively compares the performance of different Mo-based material sensors, and based on existing reviews, it deeply analyzes their working principles, electrochemical properties, application prospects, and research directions, which is of great significance for advancing the development of this field.

2. Working Principle of Mo-based Non-enzymatic Glucose Sensors

Fig. 1. shows a classical electrocatalytic model, the Pletcher model, also known as the concentric adsorption theory, which is an important theoretical model used to describe the chemical adsorption process on the electrode surface in electrocatalytic reactions[12]. This model introduces the concept of adjacent adsorption sites, unlike traditional adsorption models where adsorption sites are assumed to be uniformly independent. It suggests that the distribution of adsorption sites on the electrode surface is hierarchical, with sites at different positions having different adsorption energies. Adsorbate species first occupy the outermost sites with the lowest energy and gradually move to the inner layers, and the interactions between adjacent sites influence the behavior of the adsorbed species. This allows researchers to better understand the working principle of molybdenum-based non-enzymatic glucose sensors. In glucose sensors, Mo-based materials typically serve as catalysts or catalytic supports to enhance the efficiency of glucose oxidation. The surface of Mo-based materials can adsorb glucose molecules, providing reaction sites for the glucose oxidation process.

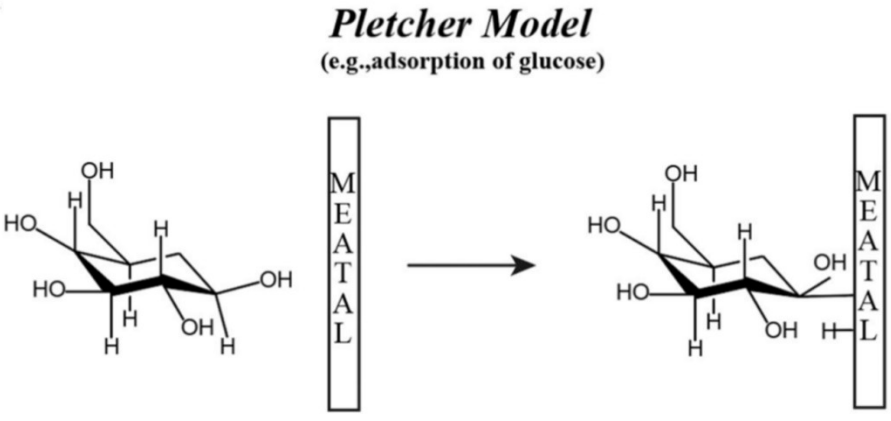

Fig. 1. Pletcher electrocatalytic mechanism diagram[12]

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Specifically, during the reaction process, the oxidation state of Mo changes, typically from a lower oxidation state (Mo^{4+}) to higher oxidation states (Mo^{5+}, Mo^{6+}). The aldehyde group on the C-position of the glucose molecule is oxidized, converting into gluconic acid. Oxygen reacts with glucose, ultimately generating hydrogen peroxide (H_2O_2) and gluconic acid, and Mo returns from the higher oxidation state to the lower oxidation state. For transition metal-based materials like Mo, as shown in Fig. 2, the mainstream mechanism of electrocatalytic glucose oxidation occurs through redox reactions at the metal center. Essentially, these materials utilize redox couples as electron transfer mediators, where the changes in the oxidation state of the transition metal lead to the further hydrolysis of glucolactone into gluconic acid, followed by the cleavage or desorption of chemical adsorption bonds[13].The section headings are in boldface capital and lowercase letters. Second level headings are typed as part of the succeeding paragraph (like the subsection heading of this paragraph). All manuscripts must be in English, also the table and figure texts, otherwise we cannot publish your paper. Please keep a second copy of your manuscript in your office. When receiving the paper, we assume that the corresponding authors grant us the copyright to use the paper for the book or journal in question. When receiving the paper, we assume that the corresponding authors grant us the copyright to use the paper for the book or journal in question. When receiving the paper, we assume that the corresponding authors grant us the copyright to use.

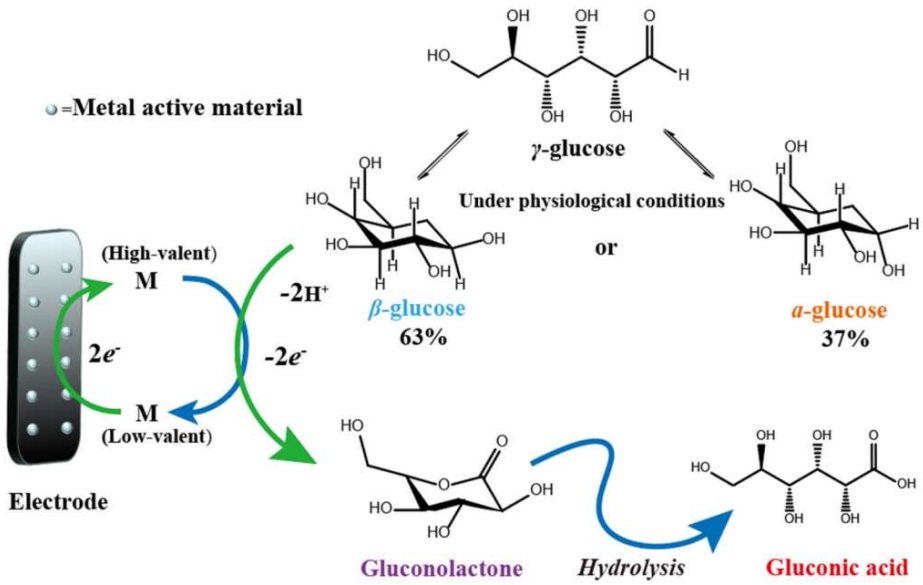

Fig. 2. Schematic diagram of electrocatalytic glucose oxidation reaction mechanism for transition metal-based non-enzymatic glucose sensors [14]

3. Research Progress on Mo-based Non-enzymatic Glucose Sensors

3.1 MoS_2-based Non-enzymatic Glucose Sensor

Mo-based materials play an important role in non-enzymatic glucose sensors, mainly including MoS_2, MoO_3, Mo atom doping, $MoSe_2$, Mo-based composites, and other forms, each with its unique properties and advantages. MoS_2 is a transition metal dichalcogenide, exhibiting typical two-dimensional material characteristics. It is composed of alternating layers of molybdenum and sulfur atoms, with each MoS_2 layer consisting of a molybdenum atomic layer sandwiched between two sulfur atomic layers, forming a stable hexagonal lattice structure. When applied to non-enzymatic glucose sensors, MoS_2 can serve as a catalytic support material, either in composite with metal nanomaterials or with metal oxide materials, to synergistically catalyze glucose oxidation reactions. The large specific surface area of MoS_2 provides abundant active sites, and it is less susceptible to environmental factors such as pH and temperature, thus maintaining good performance over a longer

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period. Huang, Dong, and others [15] synthesized Cu-MoS_2 on a GCE electrode for non-enzymatic glucose sensors, using MoS_2 as a catalytic support material, achieving a sensitivity of 1055.2 μA mM^{-1} cm^{-2} and a linear detection limit of 0-4 mM. Huang, He, and others [16] prepared a Ni-MoS_2/GCE electrode, achieving a sensitivity of 1824.4 μA mM^{-1} cm^{-2} and a detection limit of 0.31 μM (S/N=3).

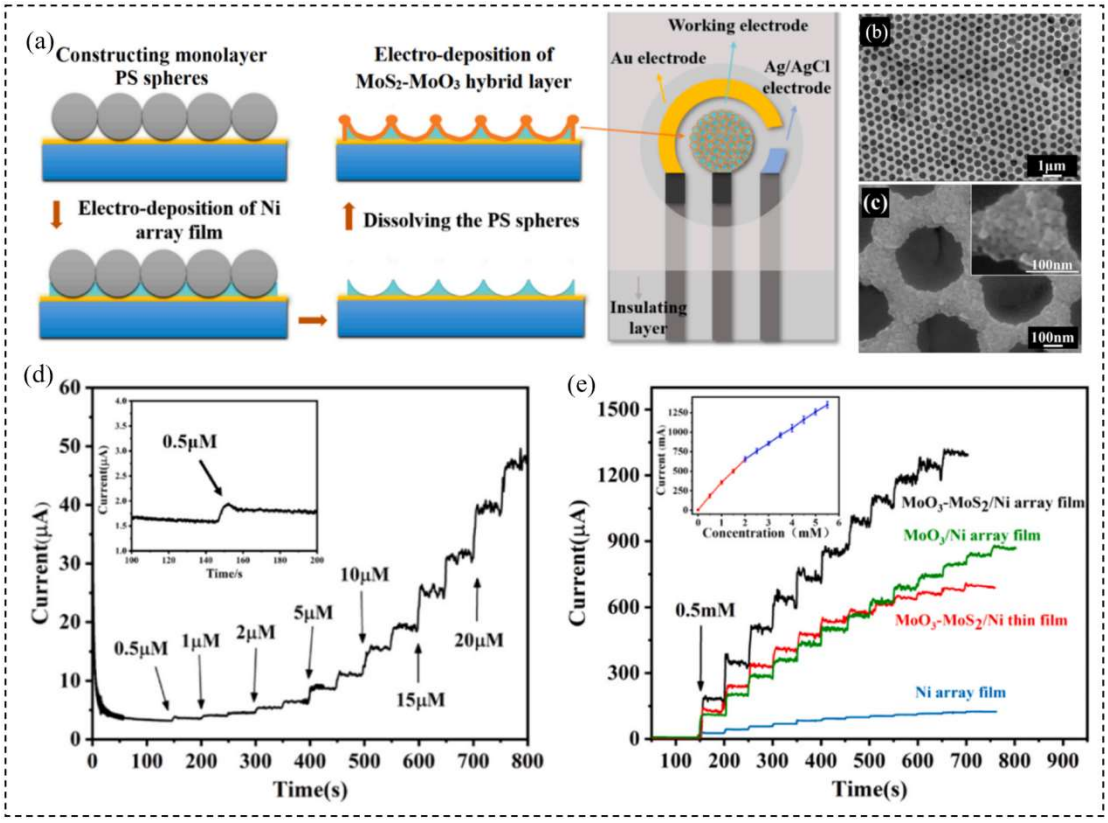

Fig. 3. (a) Schematic diagram of the preparation of MoO_3-MoS_2/Ni porous array; (b, c) SEM images of MoO_3-MoS_2/Ni; (d) Continuous addition of glucose at different concentrations to the MoO_3-MoS_2/Ni/SPE electrode at a voltage of 0.64 V in 0.2 M NaOH solution; (e) Linear relationship between the catalytic current and concentration of MoO_3-MoS_2/Ni/SPE electrode at the optimal potential. [17]

MoS_2 can also synergistically catalyze glucose oxidation reactions with metal oxides like MoO_3 and conductive polymer materials such as PPY. As shown in Fig. 3(a-c), Luo et al. [17]successfully prepared MoO_3-MoS_2/Ni porous nanoarrays on screen-printed electrodes using a two-step electrodeposition method. Due to the defects and edge-active sites of the MoO_3-MoS_2 nanostructure, the modified electrode performed well as a non-enzymatic glucose sensor. Fig. 3(d-e) show that the current response of the MoO_3-MoS_2/Ni porous array-modified SPE electrode is highly sensitive to changes in glucose concentration when different concentrations of glucose are continuously added. The calculated sensitivity is 2278 μA mM^{-1} cm^{-2}. Compared to the other three electrodes, the MoO_3-MoS_2/Ni/SPE electrode is more sensitive and exhibits better electrocatalytic performance. The calculated detection limit is 0.2 μM (S/N=3). Additionally, MoS_2 can also synergistically catalyze glucose oxidation reactions when combined with conductive polymers like polypyrrole (PPY). Ma et al. [18]prepared MoS_2-PPY-AuNPs, with a linear detection range of 0.1-80 nM and a detection limit of 0.08 nM (S/N=3). Table 1 shows various MoS_2-based non-enzymatic glucose sensors.

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Table 1. Various MoS_2-based non-enzymatic glucose sensors

Sensing materials	Linear range (mM)	Detection limit	Sensitivity ($\mu A \cdot mM^{-1}cm^{-2}$)	Response time (s)	Reference
Bilayer MoS_2	30-300	0.3 μM	260.75	<2	19
CuS-MoS_2/GCE	0.1-80 nM	1.52 μM	252.71	-	20
Co@MoS_2/CNTs	-	80 nM	131.69	4.5	21
Cu_2O-MoS_2	0.01-4	1 μM	3108.87	<2	22
GCE/CNT/MoS_2/NiNPs	0.05-0.65	0.2 μM	1212	3	23

3.2 $MoSe_2$-based Non-enzymatic Glucose Sensor

$MoSe_2$ is a transition metal compound with a two-dimensional layered structure, where the nanosheets are formed by weak van der Waals forces between Se and Mo atoms. $MoSe_2$ has a relatively large space between adjacent layers and a small bandgap ranging from 0.85 to approximately 1.6 eV, making it an excellent electrocatalytic material for non-enzymatic glucose sensors. $MoSe_2$ can effectively promote the oxidation of glucose, enhancing the sensor's sensitivity, response speed, and detection accuracy. The tunability of its electronic structure allows for further optimization of its catalytic activity through doping or surface modification. Jeevanandham et al. [24]synthesized $MoSe_2$ nanosheets decorated with nickel oxide (NiO) nanorods via a hydrothermal method using sodium molybdate and selenium metal powder. The linear detection range was 0.05-15.5 mM, with a detection limit of 0.6 μM (S/N=3).

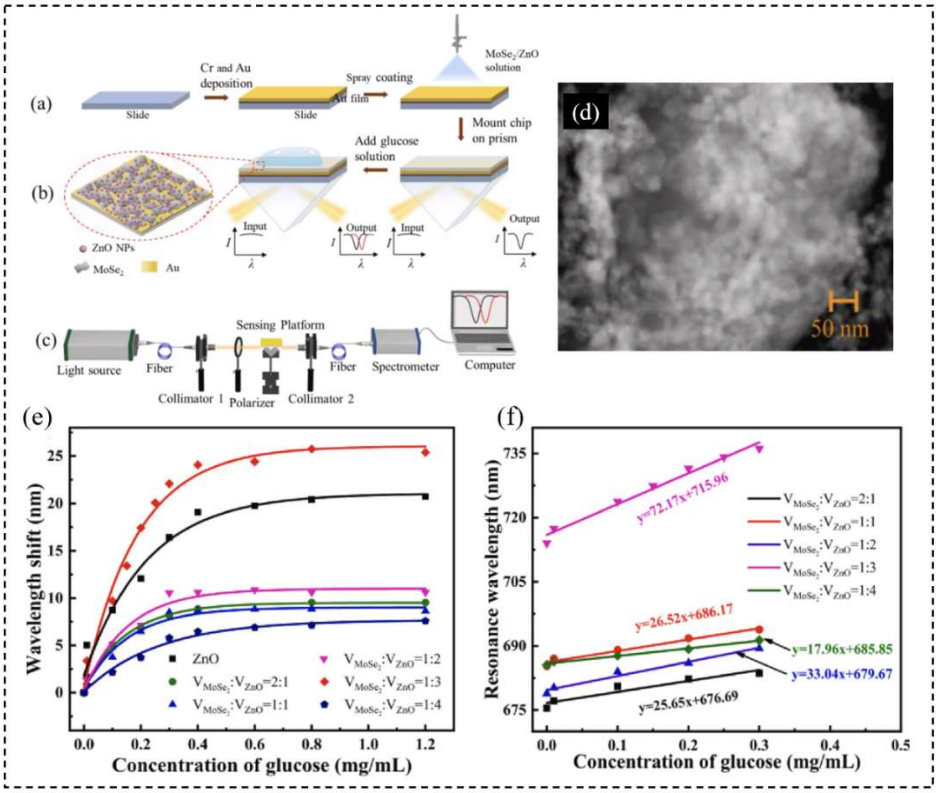

Fig. 4. (a) Schematic of the preparation of $MoSe_2$/ZnO-based SPR chip; (b) Measurement method; (c) Glucose testing device; (d) TEM image of $MoSe_2$/ZnO composite film; (e) Nonlinear fitting curve of sensitivity for sensors with different volume ratios of ZnO/$MoSe_2$ composite films; (f) Linear fitting curve of sensitivity for sensors with different volume ratios of ZnO/$MoSe_2$ composite films. [25]

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Rapid and accurate detection of glucose concentration has been shown to be crucial for human health, especially in detecting glucose at low concentrations with high precision. Chen et al. [25] studied a surface plasmon resonance (SPR) sensor for non-enzymatic glucose detection, using ZnO nanoparticles and $MoSe_2$ nanosheets composite material ($MoSe_2$/ZnO) as the sensing film, which exhibited high sensitivity and good selectivity for glucose. As shown in Fig. 4(a)(b), different volume ratios of $MoSe_2$/ZnO composites were first modified onto the gold film surface to prepare various sensors. The practical feasibility of the proposed SPR sensor for non-enzymatic glucose sensing was also studied using Kretschmann's attenuated total reflection (ATR), as shown in Fig 4(c). As illustrated in Fig. 4(e)(f), all sensors functioned within the glucose concentration range of 0–6.6 mM, with a linear increase in the range of 0–1.67 mM, reaching the optimal concentration before finally saturating. The SPR sensor with a $MoSe_2$/ZnO volume ratio of 1:3 exhibited the highest sensitivity of 72.17 nm/(mg/mL), which is a 68.62% increase compared to the sensitivity of the bare ZnO film-based sensor, which was 42.8 nm/(mg/mL). The $MoSe_2$ layer has a large surface area and high loading capacity, which helps carry a large number of glucose molecules, thus improving the immobilization efficiency of the SPR sensor. On the other hand, $MoSe_2$ has a high electron mobility, which can enhance the charge transfer rate between ZnO and glucose.

3.3 MoO_3-based Non-enzymatic Glucose Sensors

As a molybdenum-based compound, MoO_3 can promote the oxidation reaction of glucose and enhance the current response of the sensor, allowing it to maintain high sensitivity even at low glucose concentrations. Additionally, by optimizing the surface structure of MoO_3 (such as through doping or surface modification), its catalytic activity and the sensor's response speed can be further improved. Azharudeen et al. [26] synthesized MoO_3 using a sol-gel method and modified MoO_3 with PVP. The prepared PVP-modified MoO_3 nanocomposite was used as a non-enzymatic glucose sensor in a 0.1 M NaOH solution at pH 12 at room temperature, with a detection limit of 0.022 μM (S/N=3). Ren et al.[27] combined carbon materials with MoO_3 for a non-enzymatic glucose sensor, with a linear detection range of 1-100 μM, which can accurately detect glucose concentration at low glucose levels. Salarizadeh et al. [28]synthesized NiO-MoO_3 nanocomposites via a hydrothermal method for glucose detection, achieving a detection limit of 0.85 μM (S/N=3). Using the chronoamperometry method, this sensor maintained 65% of its initial current, demonstrating excellent stability. Table 2 shows different MoO_3-based non-enzymatic glucose sensors.

Table 2. Different MoO_3-based Non-enzymatic Glucose Sensors

Sensing materials	Linear range (mM)	Detection limit	Sensitivity ($\mu A \cdot mM^{-1}cm^{-2}$)	Reference
MoO_3/GCE	0.005- 0.175	51 μM	15.4	29
NiO–MoO_3/GCE	0.2-1	0.85 μM	-	30
PF-MoO_3/GCE	0.277-0.972	0.079 mM	42.9	31
MoO_3/NiO	5–175,175–615	51.07,104.17 mM	15.4,7.5	32
MoO_3-MoS_2/Ni	0.5 μM–2 mM	0.2 μM	2278	17

3.4 Mo Atom Doping, Mo-Based Composites

Molybdenum (Mo) atoms can be introduced into other materials to improve their properties, thereby enhancing the sensitivity, selectivity, and stability of non-enzymatic glucose sensors. Zhang et al. [33]co-deposited Ni-Mo-Nb metallic glass films on a three-dimensional printed polymer scaffold for glucose detection. The composite material combines the high electrochemical activity of Ni-Mo-Nb amorphous alloys with the stability provided by the micro-nanostructure, resulting in high sensitivity (482 μA mM^{-1} cm^{-2}), a wide linear range (0-1 mM and 1-5 mM), a low detection limit (2.94 μM

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(S/N=3)), and excellent long-term stability and selectivity in alkaline solutions. As shown in Figure 5, Ravitchandiran et al. [34] prepared ZnFe(PBA)@Mo_3C_2Tx for glucose sensors using an in situ ultrasound method. The sensor exhibited a sensitivity of 818.2 μA mM^{-1} cm^{-2} and a detection limit of 1.6 μM (S/N=3). The sensor was used to analyze glucose in human sweat samples, and the relative standard deviation (RSD) of the obtained results compared to the corresponding blood glucose values was 2.98%.

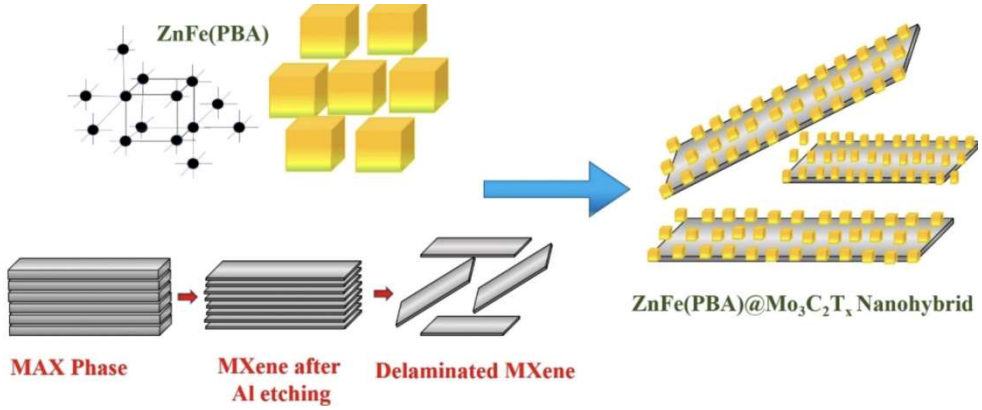

Fig. 5. Schematic of the Preparation of ZnFe(PBA)@Mo_3C_2Tx Hybrid Material[34]

4. Application Prospects of Mo-based Non-enzymatic Glucose Sensors

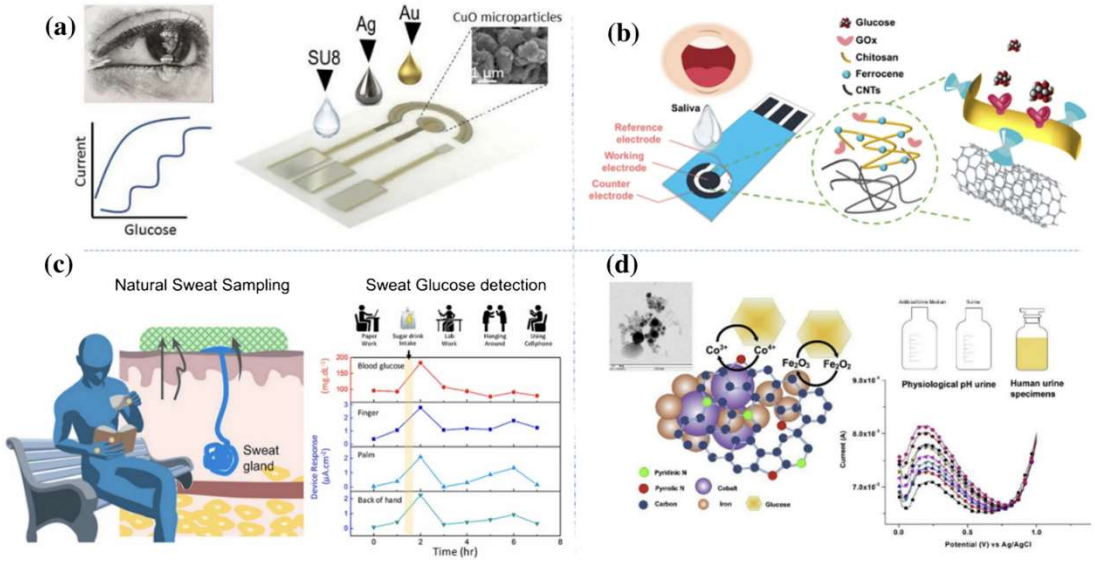

Fig. 6. (a) Non-enzymatic glucose sensor for tear analysis [35]; (b) Non-enzymatic glucose sensor for saliva analysis using screen-printed electrodes[36]; (c) Non-enzymatic glucose sensor for natural sweat detection [37]; (d) Blood glucose detection using human urine as a sample [38].

Mo-based non-enzymatic glucose sensors have broad application prospects in fields such as healthcare, wearable devices, food monitoring, and environmental protection. In the healthcare sector, they can provide accurate and rapid blood glucose monitoring, especially for diabetes management[39]. Molybdenum-based glucose sensors are expected to develop towards miniaturization and non-invasive detection, enabling their integration into smart wearable devices. As shown in Fig. 6 (a-d), non-enzymatic glucose sensors can determine glucose concentrations by detecting biological fluids such as tears, saliva, sweat, and urine, improving safety, comfort, and efficiency in testing. In the food industry, glucose sensors can be used to monitor sugar content in real-time, ensuring food safety[40]. In environmental monitoring, they can detect glucose concentrations in water, assess

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pollution levels, or track biodegradation processes. Additionally, molybdenum-based sensors can be applied in smart drug delivery systems, adjusting drug release based on real-time blood glucose data to provide personalized treatment.

5. Conclusion

Mo-based non-enzymatic glucose sensors have shown tremendous application potential in various fields, such as blood glucose monitoring, smart health devices, food safety testing, and environmental monitoring, due to their low cost, high stability, excellent electrocatalytic performance, and strong environmental adaptability. Mo-based materials, including MoS_2, MoO_3, $MoSe_2$, Mo atom doping, and their composites, have played a crucial role in enhancing sensor sensitivity, response speed, and durability. Especially when compared to precious metals and carbon-based materials, Mo-based materials exhibit more economical and stable advantages. Through appropriate surface modification and doping, the catalytic activity of Mo-based materials can be further optimized, thereby improving their performance in complex environments.

Despite the significant progress made in Mo-based non-enzymatic glucose sensors, challenges remain in improving the sensor's sensitivity and selectivity, enhancing long-term stability, and reducing production costs. Future research will focus on the design of multifunctional composite materials, miniaturization and integration of sensors, and the improvement of non-invasive detection capabilities for various biological samples (such as sweat, saliva, etc.). With continuous innovation and optimization of related technologies, it is believed that the performance of Mo-based non-enzymatic glucose sensors will further improve and achieve breakthroughs in commercial applications, becoming an important technology in the future of health monitoring and environmental protection.

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